

CLAIMS

What is claimed is:

1. A solid multi-component membrane for use in an electrochemical reactor characterized by (1) an intimate, gas-impervious, multi-phase mixture of an electronically-conductive phase and an oxygen ion-conductive phase or (2) a mixed metal oxide material having a perovskite structure represented by the formula:



wherein A represents a lanthanide or Y, or a mixture thereof; A' represents an alkaline earth metal or a mixture thereof; B represents Fe; B' represents Cr or Ti, or a mixture thereof; and B'' represents Mn, Co, V, Ni or Cu, or a mixture thereof and s, t, u, v, w, and x each represent a number such that:

s/t equals from about 0.01 to about 100;

u equals from about 0.01 to about 1;

v equals from 0.01 to about 1;

w equals from zero to about 1;

x equals a number that satisfies the valences of the A, A', B, B' and B'' in the formula; and

$$0.9 < (s+t)/(u+v+w) < 1.1.$$

2. The solid membrane of claim 1 comprising an intimate, gas-impervious multi-phase mixture (1) wherein the electronically-conductive phase comprises silver, gold, platinum, palladium, rhodium, ruthenium, bismuth oxide, a praseodymium-indium oxide mixture, a cerium-lanthanum oxide mixture, a niobium-titanium oxide mixture, or an electron-conductive mixed metal oxide of a perovskite structure, or a mixture thereof and the oxygen ion-conductive phase comprises yttria- or calcia-stabilized zirconia, ceria or bismuth oxide, or an oxygen ion-conductive mixed metal oxide of a perovskite structure.

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3. The solid membrane of claim 1 comprising an intimate, gas-impervious multi-phase mixture (1) prepared by a process which comprises the steps of

(A) preparing an intimate multi-phase mixture of at least one material which is electronically-conductive and at least one oxygen ion-conductive material,

(B) forming the mixture into a desired shape, and

(C) heating the formed mixture to a temperature of at least about 500°C to form a dense and solid membrane.

4. The solid membrane of claim 1 comprising an intimate, gas-impervious multi-phase mixture (1) prepared by the process which comprises the steps of

(A) preparing an intimate multi-phase mixture of at least one metal oxide, the metal of which is electronically-conductive, and at least one oxygen ion-conductive material,

(B) heating the mixture at an elevated temperature in a reducing atmosphere to reduce the metal oxide to metal,

(C) forming the reduced mixture into a desired shape, and

(D) heating the formed mixture to a temperature of at least about 500°C to form a dense and solid membrane.

5. The solid membrane defined according to claim 1 wherein the solid membrane comprises an intimate, gas-impervious, multi-phase mixture consisting essentially of from about 1 to about 75 parts by volume of the electronically-conductive phase and from about 25 to about 99 parts by volume of the ionically-conductive phase.

6. The solid membrane of claim 1 comprising a mixed metal oxide material having a perovskite structure (2) wherein A represents La or Y, or a mixture thereof; A'

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represents Ca or Sr, or a mixture thereof; B' represents Cr; and B'' represents Mn or Co or a mixture thereof;

s/t equals from about 0.1 to about 20;

v equals from about 0.05 to about 0.5;

w equals from about 0.01 to about 0.5.

7. An element for use in an electrochemical reactor or reactor cell having a first surface capable of reducing oxygen to oxygen ions, a second surface capable of reacting oxygen ions with an oxygen-consuming gas, an electron-conductive path between the first and second surfaces and an oxygen ion-conductive path between the first and second surfaces characterized in that the element comprises a sulfur reducing catalyst at the first surface of the element.

8. An element for use in an electrochemical reactor or reactor cell having a first surface capable of reducing oxygen to oxygen ions, a second surface capable of reacting oxygen ions with an oxygen-consuming gas, an electron-conductive path between the first and second surfaces and an oxygen ion-conductive path between the first and second surfaces characterized in that the element comprises (A) a solid multi-component membrane characterized by (1) an intimate, gas-impervious, multi-phase mixture of an electronically-conductive phase and an oxygen ion-conductive phase or (2) a mixed metal oxide material having a perovskite structure and (B) a conductive coating, a catalyst, or a conductive coating comprising a catalyst.

9. The element of claim 8 wherein (B) is a conductive coating comprising an alkali or alkaline earth metal or metal oxide in an amount in the range from about 1% to about 50% by weight of the conductive coating.

10. An electrochemical reactor cell for reacting an oxygen-consuming gas with an oxygen-containing gas in an environment containing either the oxygen-consuming gas or

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the oxygen-containing gas, characterized by a solid multi-component membrane having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, wherein the solid membrane is defined according to any one of claims 1-6.

11. The electrochemical reactor cell of claim 10 wherein the solid membrane is a solid cylindrical core having a circular passage for the movement of one or more gases therethrough.

12. An electrochemical reactor cell for reacting an oxygen-consuming gas with an oxygen-containing gas in an environment containing either the oxygen-consuming gas or the oxygen-containing gas, characterized by an element having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, wherein the element is defined according to any one of claims 7-9.

13. An electrochemical reactor for reacting an oxygen-consuming gas with an oxygen-containing gas characterized by:

a shell having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, and

at least one electrochemical reactor cell positioned within the shell having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, so that the shell and the reactor cell together form a first zone for introducing, reacting and expelling a first gas or gas mixture and the passage through the reactor cell forms a second zone within the electrochemical reactor for introducing, reacting and expelling a second gas or gas mixture,

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wherein the electrochemical reactor cell comprises a solid multi-component membrane as defined in any one of claims 1-6.

14. An electrochemical reactor for reacting an oxygen-consuming gas with an oxygen-containing gas characterized by:

a shell having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, and

at least one electrochemical reactor cell positioned within the shell having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, so that the shell and the reactor cell together form a first zone for introducing, reacting and expelling a first gas or gas mixture and the passage through the reactor cell forms a second zone within the electrochemical reactor for introducing, reacting and expelling a second gas or gas mixture,

wherein the electrochemical reactor cell comprises an element as defined in any one of claims 7-9.

15. The electrochemical reactor of claim 13 wherein the electrochemical reactor comprises a catalyst on a support in the first zone.

16. The electrochemical reactor of claim 14 wherein the electrochemical reactor comprises a catalyst on a support in the first zone.

17. An electrochemical reactor for reacting an oxygen-consuming gas with an oxygen-containing gas characterized by:

a shell having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, and

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at least one electrochemical reactor cell positioned within the shell having an entrance end, an exit end and a passage therebetween for the movement of one or more gases from the entrance end to the exit end, so that the shell and the reactor cell together form a first zone for introducing, reacting and expelling a first gas or gas mixture and the passage through the reactor cell forms a second zone within the electrochemical reactor for introducing, reacting and expelling a second gas or gas mixture,

wherein the electrochemical reactor comprises an oxidative coupling catalyst on a support in the first zone.

18. An electrochemical process for oxidizing a gas capable of reacting with oxygen characterized by

(A) providing an electrochemical reactor cell comprising first and second zones separated by a solid multi-component membrane as defined according to any one of claims 10-12,

(B) heating the electrochemical cell to a temperature of from about 300°C to about 1400°C,

(C) passing an oxygen-containing gas in contact with the membrane in the first zone, and

(D) passing a gas capable of reacting with oxygen in contact with the membrane in the second zone.

19. An electrochemical process for oxidizing a gas capable of reacting with oxygen characterized by

(A) providing an electrochemical reactor cell comprising first and second zones separated by an element as defined according to claim 8 or 9,

(B) heating the electrochemical cell to a temperature of from about 300°C to about 1400°C,

(C) passing an oxygen-containing gas in contact with the membrane in the first zone, and

(D) passing a gas capable of reacting with oxygen in contact with the membrane in the second zone.

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20. The process of claim 18 or 19 conducted at a temperature of about 1000°C to about 1400°C, wherein the gas capable of reacting with oxygen is methane, natural gas, ethane, or other light hydrocarbon gas, or a mixture thereof, and further comprising

(E) recovering synthesis gas from the second zone.

21. The process of claim 20 wherein (D) comprises passing a methane-steam mixture in contact with the membrane in the second zone.

22. The process of claim 18 or 19 conducted at a temperature of about 550°C to about 950°C, wherein the gas capable of reacting with oxygen is methane, natural gas, ethane, or other light hydrocarbon gas, or a mixture thereof, and further comprising

(E) recovering one or more unsaturated hydrocarbons from the second zone.

23. An electrochemical process for producing unsaturated hydrocarbon compounds from saturated hydrocarbon compounds characterized by:

(A) providing an electrochemical cell comprising an element having a first and second surface as defined above;

(B) passing an oxygen-containing gas in contact with the first surface while

(C) passing a saturated hydrocarbon-containing gas in contact with a dehydrogenation catalyst adjacent to the second surface and

(D) recovering unsaturated hydrocarbons

wherein the element is defined as in claim 8 or 9 or comprises a solid multi-component membrane as defined in any one of claims 1-6.

24. An electrochemical process for substitution of aromatic compounds which comprises:

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(A) providing an electrochemical cell comprising  
(i) an element having a first surface capable of reducing oxygen to oxygen ions, a second surface capable of reacting oxygen ions with an oxygen-consuming gas, an electron-conductive path between the first and second surfaces and an oxygen ion-conductive path between the first and second surfaces,

(ii) a first passageway adjacent to the first surface, and

(iii) a second passageway adjacent to the second surface; and

(B) passing an oxygen-containing gas through the first passageway while

(C) passing an oxygen-consuming gas through the second passageway wherein the oxygen-consuming gas comprises a mixture of a hydrogen-containing aromatic compound and a second hydrogen-containing compound to produce the substituted aromatic compound.

25. A process for extracting oxygen from  $N_2O$ ,  $NO$ ,  $NO_2$ ,  $SO_2$  and/or  $SO$ , characterized by the process of claim 18 wherein step (C) comprises passing a gas containing  $N_2O$ ,  $NO$ ,  $NO_2$ ,  $SO_2$ , or  $SO$ , or a mixture thereof, in contact with the solid membrane in the first zone.

26. A process for extracting oxygen from  $N_2O$ ,  $NO$ ,  $NO_2$ ,  $SO_2$  and/or  $SO$ , characterized by the process of claim 19 wherein step (C) comprises passing a gas containing  $N_2O$ ,  $NO$ ,  $NO_2$ ,  $SO_2$ , or  $SO$ , or a mixture thereof, in contact with the element in the first zone.

27. The process of claim 25 or 26 conducted at a temperature in the range from about  $300^\circ C$  to about  $1000^\circ C$ .

28. An electrochemical process for producing hydrogen cyanide from methane and ammonia characterized by

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(A) providing an electrochemical reactor cell comprising first and second zones separated by an element, having a first surface capable of reducing oxygen to oxygen ions, a second surface capable of reacting oxygen ions with an oxygen-consuming gas, an electron-conductive path between the first and second surfaces and an oxygen ion-conductive path between the first and second surfaces;

(B) heating the electrochemical reactor cell to a temperature of from about 1000°C to about 1400°C;

(C) passing an oxygen-containing gas in contact with the element in the first zone;

(D) passing methane and ammonia in contact with the membrane in the second zone; and

(E) recovering hydrogen cyanide from the second zone.

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